



Estimating soil nitrate leaching of nitrogen fertilizer from global meta-analysis

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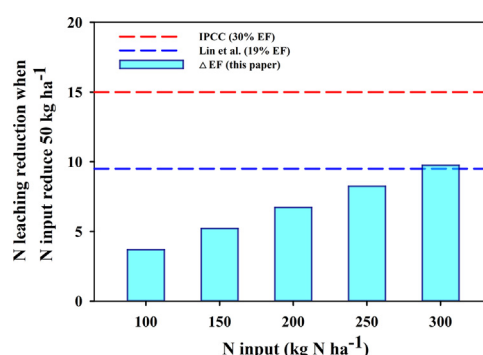
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HIGHLIGHTS

- The estimation using constant EF covered the effect of excessive fertilization on nitrate leaching.
- Fertilizer-induced EFs of nitrate leaching are not constant but increase with N additions.
- Δ EF model provided a more accurate empirical model for nitrate leaching worldwide.

GRAPHICAL ABSTRACT



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ABSTRACT

Global estimates of soil nitrate leaching of applied nitrogen (N) in agricultural production systems are not imprecise; however, the results of some field experiments have suggested that nitrate leaching responds exponentially rather than linearly to increasing N inputs. In this study, we compiled field data on nitrate leaching for 324 site-year combinations extracted from 86 peer-reviewed articles to test the hypothesis that in response to N fertilizer addition, soil nitrate leaching emission factors (EF) do not remain constant, but rather increase rapidly with increasing rates of N application. The averaged overall results showed that the proportional change in the EF response to increasing N input (Δ EF, %) was 0.042. Because this value was positive and significantly different from zero, our results demonstrate that EF is not constant, but rather increases with N addition. We compared our Δ EF response pattern with other constant EF patterns, and found that the 30% EF estimate of the Intergovernmental Panel on Climate Change and the 19% EF used in another published model were too high. A global estimate of soil nitrate leaching in arable uplands calculated with our Δ EF pattern was substantially lower than other estimates calculated with other procedures. In conclusion, our Δ EF pattern provided a globally applicable procedure for more accurate estimation of leaching loss and potential environmental costs incurred by various N application gradients.

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1. Introduction

Nitrogen (N) is both an essential nutrient and a major pollutant in agricultural ecosystems (Zhang et al., 2015). Countries face the joint

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challenges of food security and environmental degradation, and have invested considerably in synthetic fertilizers for the crop production system. However, anthropogenic activities continue to profoundly change the N cycle of the pedosphere, atmosphere, and hydrosphere, as they have since the industrial revolution (Howarth et al., 1996; Galloway et al., 2004). In particular, modern intensive agricultural production systems are always associated with substantial reactive N loss through hydrological and gaseous pathways (Tilman et al., 2011; Burney et al., 2010; Quemada et al., 2013). Among these N loss pathways, nitrate leaching is considered to be the dominant cause of rising nitrate concentrations in groundwater, and directly responsible for ecosystem eutrophication and water quality degradation (Rivett et al., 2008; Robertson and Vitousek, 2009; Zhou et al., 2012; Sebilo et al., 2013).

Soil is the major source of nitrate leaching, and the total input of N to soil can be a robust predictor to evaluate nitrate leaching intensity. A fertilizer-induced emission factor (EF) has been applied extensively (Intergovernmental Panel on Climate Change [IPCC]; Ju et al., 2009; Cui et al., 2018) as a representative value relating the quantity of a pollutant in a substrate to the activities controlling its release into that substrate. In recent decades, the IPCC has used the EF approach to estimate global losses of nitrate from fertilized agricultural systems (IPCC, 2006). Under this approach, nitrate N lost by leaching is assumed to be a set proportion of applied fertilizer N, typically 30%. Statistical models or integrated data analyses based on field tests have also been developed; these indicated that the global EF was likely a constant (Boy-Roura et al., 2016; Lin et al., 2001; Ma et al., 2012; Zhou and Butterbach-Bahl, 2014). For example, Lin et al. (2001) performed a simulation with a global process-based model, and showed that the EF of nitrate leaching was 19% of the total N fertilizer input. A global meta-analysis based on 205 observations extracted from 32 studies attempted to provide insight into nitrate leaching in maize and wheat cropping systems (Zhou and Butterbach-Bahl, 2014).

Most analyses have considered the EF value as a constant that spans the contribution of different fertilizer gradients to N leaching (IPCC; Sorensen and Rubaek, 2012). Recent field experiments have shown that the EF is not influenced by biological thresholds, including the point at which N input exceeds crop demand. These experiments indicated that little nitrate would leach if N fertilizer application were less than that required for optimum or maximum crop yield. They also showed that leaching would increase sharply once the N fertilizer rate exceeded the optimum level (Goulding et al., 2000; Delin and Stenberg, 2014; Ruan et al., 2016; Perego et al., 2012). The non-linear nitrate leaching response to increasing N inputs should be tested across a range of fertilizer formulations, climates, and soil types because the vast majority of studies on nitrate leaching have examined a single fertilizer at a specific site.

Across global agricultural systems, calculations of national nitrate leaching rates based on a constant EF cannot accurately represent the leaching status of the individual countries. Agricultural N balances are widely variable among nations, depending on economic development. An input deficit of N cannot maintain soil fertility in some developing regions, but excessive N surpluses can even lead to environmental damage in some rapidly developing countries (Vitousek et al., 2009). For example, on the North China Plain, a typical high-input region, the N fertilization rate exceeds $400 \text{ kg N ha}^{-1} \text{ year}^{-1}$ in maize/wheat rotations. Excessive N input exceeds crop demand, and the N surplus can reach 150 kg N ha^{-1} , causing N loss to the environment via multiple pathways (Chen et al., 2014; Cui et al., 2014). In these regions of excessive N fertilization, incorporating an exponential EF into leaching loss models would maintain a higher-level correspondence with high N input, avoiding an underestimation of the environmental costs of fertilizer inputs.

In this study, we conducted a meta-analysis to estimate the nonlinear response of nitrate leaching to N application rates using the ΔEF method developed by Shcherbak et al. (2014) to characterize the

nonlinear N_2O response to N input. We also examined several potential factors influencing ΔEF , including crop type, fertilizer type, and other available experimental factors. We compared the ΔEF pattern determined in this study with those reported in other publications, such as an IPCC report that used a constant EF value of 30%. Finally, we applied a nonlinear fitting function to quantitatively evaluate and determine the nitrate leaching levels on arable uplands.

2. Materials and methods

2.1. Database compilation

We conducted a comprehensive literature search of peer-reviewed publications that reported fertilizer-induced nitrate leaching from soils prior to October 2018. All data were derived from publications found by searching for key words ('nitrate leaching', 'nitrogen fertilizer', 'N input' and 'soil') in the Web of Science database and the China Knowledge Resource Integrated Database. We also scrutinized publications cited in the meta-analyses of Zhou and Butterbach-Bahl (2014) and Quemada et al. (2013).

We followed the method developed by Shcherbak et al. (2014) and included studies satisfying the following criteria. First, we included only field-experiment nitrate leaching data, excluding laboratory or simulation results. Second, all nitrate leaching experiments contained at least three N input levels including a zero N control. Third, cumulative nitrate leaching during the crop growth period had to be explicitly reported with the published data. If the data were presented in a graph, we digitized the data using the Get Data Graph Digitizer. Finally, we included only reports that measured N leaching using lysimeter and suction cup methodologies (Quemada et al., 2013). For accuracy, our analysis required the following details in the selected publications: location (longitude/latitude), climatic conditions (climate type, annual mean temperature, and precipitation), soil properties (soil pH, soil organic carbon [SOC], and total N [TN]), crop types, fertilization management procedures (fertilizer types, N application rate, and frequency of fertilization), total soil nitrate leaching, and measurement procedures. We included 86 peer-reviewed papers spanning 324 site-years in this meta-analysis; the distribution of all locations is shown in Fig. 1. All 324 site-year ΔEFs were classified into categories based on experimental factors: crop type (upland crops, including maize and wheat, rice, grasses, and vegetables), fertilizer type (urea, controlled-release fertilizer [CRF] containing controlled-release compound fertilizer and controlled-release urea, ammonium nitrate [AN], and manure), and other experimental factors (SOC, soil TN, soil pH, mean annual precipitation, fertilizer application frequency, and measurement procedures (including lysimeter and suction cup methodologies). The soil nutrient categorization standard of Lv and Li (2006) provided the classification criteria for SOC, TN, and pH. We also used the classification of Shcherbak et al. (2014) to provide criteria for the categorization of precipitation and fertilization frequency (Shcherbak et al., 2014). We selected 1%, 1 g kg^{-1} , 7.0, 700 mm, and 1 as the dividing thresholds for categories of SOC, TN, pH, precipitation, and fertilization frequency, respectively. We recognized two procedures for measuring N leaching: the lysimeter and suction cup methodologies. Across the data for 324 site-years, the lysimeter method accounted for 70.7% of measurements, and the suction cup method accounted for 29.3% (Table 1).

2.2. Data analyses

We aimed to apply a new methodology to determine the nonlinear increase in nitrate leaching with increasing N application rate. The ΔEF method used in this study applied the relationship between N input and nitrate leaching to calculate EF_N for every non-zero N input thus: $\text{EF}_N = (\text{ER}_N - \text{ER}_0) / N \times 100$, where ER_N is the amount of nitrate leaching for non-zero N input; ER_0 is the nitrate leaching for zero N (control), and N is the N application rate. The linear relationship

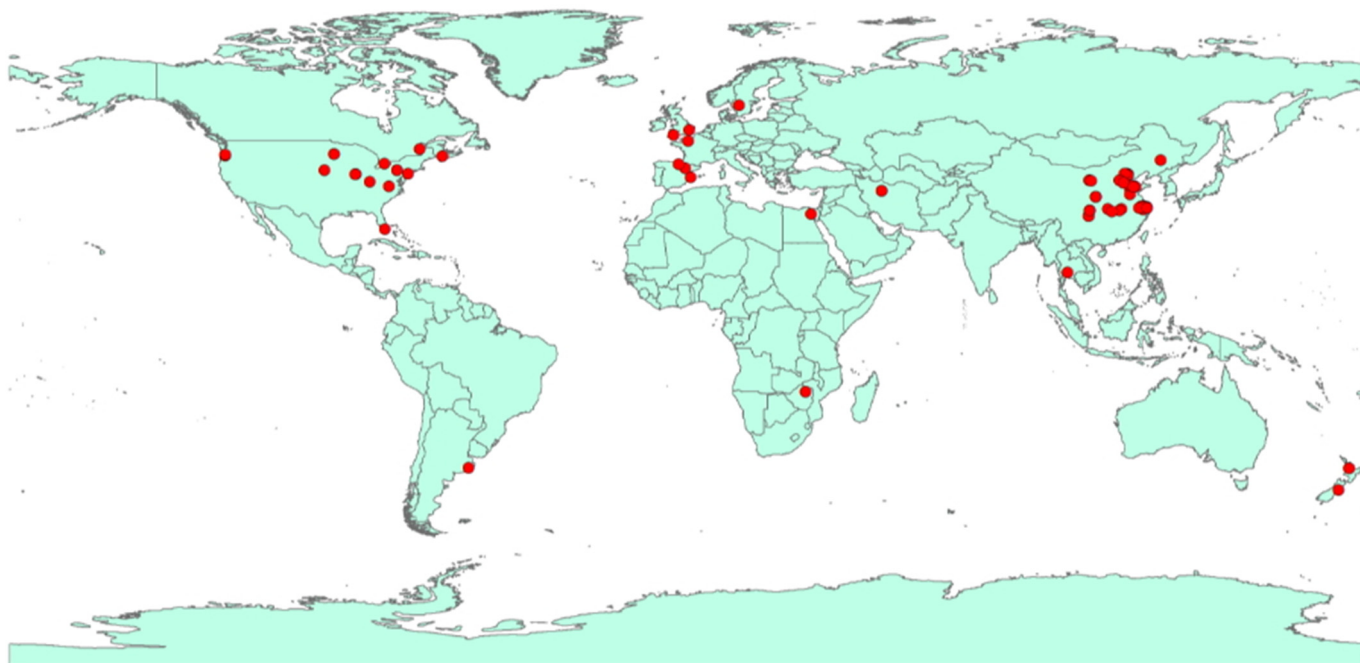


Fig. 1. Locations of sites included in this study (n = 84 sites).

between EF and the N application rate is described by the following expression: $EF_N = EF_0 + \Delta EF N$, where EF_0 and ΔEF are the intercept and slope of the effect function, respectively. We calculated ΔEF through each site-year combination containing at least three N gradients (zero, relatively low, and high N inputs). The ΔEF could be regarded as the degree of nonlinearity of the nitrate leaching response to the N rate: zero ΔEF indicates that nitrate leaching grows linearly with the N rate (constant EF), positive ΔEF indicates that nitrate leaching increases faster than a linear response with N rate (increasing EF), and negative ΔEF indicates that nitrate leaching increases slower than a linear response

(decreasing EF). For conciseness and accuracy, ΔEF in this paper is expressed as a percentage.

To accurately determine the effect of N input on nitrate leaching, we subtracted the individual check value from each nitrate leaching estimate, and used the paired net leaching and N application rates to express the upland crop quadratic model thus: $ER = 0.01 N (EF_0 + \Delta EF N)$. The current global mean nitrate leaching EF value derived from synthetic fertilizer or livestock manure is 30%, as calculated using the IPCC methodology (IPCC, 2006). We considered the IPCC default EF for nitrate leaching to be 30% and included this value in our calculations. The nitrate leaching pattern of Lin et al. (2001) was also included in the comparisons, which used a global-based process simulation model that was developed for the N cycle in terrestrial ecosystems to determine a 19% nitrate leaching EF for a global estimate.

We conducted a Kolmogorov-Smirnov test for the ΔEF datasets and found that the ΔEF distribution pattern was not normal (Fig. 2). Thus, we used nonparametric (re-sampling) and parametric procedures for

Table 1

The effects of crop type, fertilizer type, and experimental factors on mean emission factor change rates (ΔEF , %); n, number of observations; $P < 0.05$, statistically significant differences between the mean ΔEF and zero. Different letters indicate significant differences within each category between crop types and fertilizer types, and between paired levels of other experimental factors.

Category	n	Mean	SEM	P value	Group
All	324	0.042	0.007	0.000	
Crop type					
Upland crops	144	0.030	0.010	0.000	ab
Rice	30	0.005	0.002	0.003	b
Grasses	38	0.040	0.010	0.001	ab
Vegetables	112	0.069	0.013	0.000	a
Fertilizer type					
Urea	113	0.023	0.011	0.039	a
CRF	11	0.026	0.013	0.072	a
AN	39	0.077	0.023	0.002	a
Manure	32	0.034	0.005	0.000	a
Experimental factors					
Soil organic carbon <1%	108	0.030	0.010	0.003	a
Soil organic carbon ≥1%	127	0.048	0.008	0.000	a
pH < 7	110	0.062	0.012	0.000	b
pH ≥ 7	102	0.021	0.006	0.000	a
Total N < 1	76	0.029	0.013	0.033	a
Total N ≥ 1	103	0.034	0.006	0.000	a
Precipitation < 700 mm	104	0.024	0.012	0.042	a
Precipitation ≥ 700 mm	118	0.045	0.012	0.000	a
Fertilizer application = 1	52	0.063	0.017	0.001	b
Fertilizer application > 1	121	0.031	0.008	0.000	a
Lysimeter	229	0.044	0.007	0.000	a
Suction cup	95	0.038	0.016	0.019	a

CRF, controlled-release fertilizer; AN, ammonium nitrate; N, nitrogen.

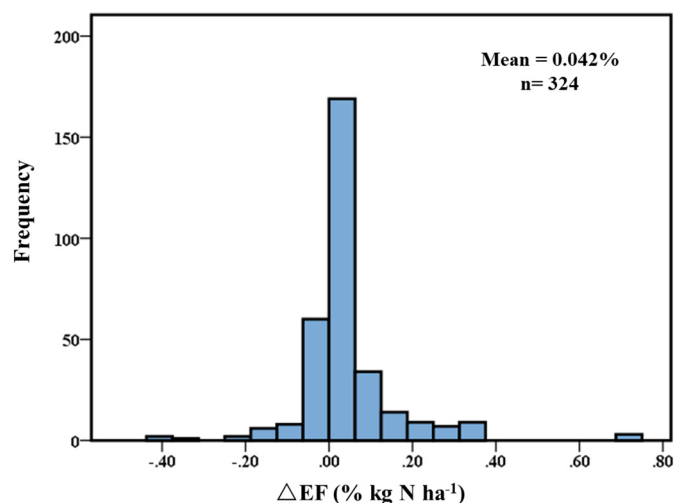


Fig. 2. Histogram of nitrate leaching emission factor change rates (ΔEF ; n = 324) with a mean value of 0.042%.

further analysis. Mean ΔEF values were calculated across categories and for each variable in each category; bias-corrected 95% confidence intervals (CIs) were generated using a bootstrapping procedure ($n = 4999$). Mean values were considered significantly different from zero when their 95% CIs did not overlap with zero and the randomization tests yielded P -values < 0.05 . We also analyzed the mean response ratios of the environmental factors (SOC, TN, soil pH, and mean annual precipitation) and management factors (crop and fertilization management, and measurement methodology) used in the field experiments. Descriptive statistical parameters of the available influencing factors were calculated to estimate the mean ΔEF of the variables and the standard errors of the means (SEMs). To identify significant differences within groups, the data were subjected to a one-way ANOVA, and significant differences between factors were identified using the least significant difference test ($P < 0.05$). In our tabular compilations (Table 1, column 6), means within the crop type and fertilizer type categories that do not share lower case letter are significantly different (ANOVA, $P < 0.05$). Different lower case letters also identify significant differences between the pairs of means within the six categories of experimental factors (Table 1).

2.3. Estimating nitrate leaching for upland crops

To estimate nitrate leaching in arable uplands, we extracted the information of total N fertilizer use for maize and wheat from the database of the International Fertilizer Industry Association (IFA; <https://www.ifastat.org/>) and cropland area data for major countries from the Food and Agriculture Organization database (FAO; <http://www.fao.org/faostat/en/#data>). N fertilizer use and arable land area in our study accounted for 83.9% and 63.4% of global totals, respectively. We calculated N input per hectare as the quotient of the total amount of N divided by the arable land area, and estimated nitrate leaching with three leaching models using the N input per hectare. The 'Country-crops' column in Table 2 indicates the numbers of major country/crop combinations included the calculations (Tables 2 and S2).

3. Results

The mean across all 324 ΔEF values (%) was 0.042, a value significantly different from zero ($P < 0.001$; Table 1), indicating an increase in nitrate leaching with N input, but the relationship was not linear. Upland crops (0.030), rice (0.005), grasses (0.040), and vegetables (0.069) all had positive ΔEF values (Table 1, $P < 0.01$). The vegetable category had the highest ΔEF , which was significantly different from that of rice, but not from those of other crop types (Table 1).

Among fertilizers, urea, ammonium nitrate, and manure all had positive mean ΔEF values ($P < 0.05$). Urea ($n = 113$) dominated the other available fertilizer category (CRF, $n = 11$; AN, $n = 39$; and manure, $n = 32$) and had the lowest, but still positive ΔEF (0.023). Although not significantly different from zero ($P = 0.072$; Table 1), the ΔEF of CRF

was positive, but low. Ammonium nitrate had a higher ΔEF value than any other fertilizer type in our study.

Among other available experimental factors, significant pairwise differences (t -test) were detected between levels for soil pH (< 7 vs. ≥ 7) and fertilizer application frequency (1 vs. > 1). However, the mean ΔEF value at a soil pH ≥ 7 was the lowest among all influencing factors, but still positive ($\Delta EF = 0.021$, $P < 0.001$; Table 1). The ΔEF s were positive and somewhat elevated within categories of influencing factors under the following conditions: SOC $\geq 1\%$, soil TN ≥ 1.0 g kg $^{-1}$, and annual precipitation > 700 mm. ΔEF estimates were not significantly different between suction cup and lysimeter methodologies.

We extracted the upland crop data for N input and nitrate leaching rate in pairs, and fitted a curve to determine a quadratic model in the form: $ER = 0.01 N (2.84 + 0.030 N)$, where ER is the nitrate leaching rate (kg NO $_3$ -N ha $^{-1}$) and N is the N fertilizer application rate (kg ha $^{-1}$) (Fig. 3A). Compared with the IPCC 30% nitrate leaching EF and another nitrate leaching pattern (19% EF) calculated from the global nitrate leaching rate through model simulation by Lin et al. (2001), our estimated ΔEF values were lower across a range of N fertilizer rates (0–500 kg N ha $^{-1}$) (Fig. 3B). Our ΔEF model showed an increase in N leaching with increasing N application rates. Thus, our ΔEF model calculated that EF increased from 6% at 100 kg N ha $^{-1}$ to 12% at 300 kg N ha $^{-1}$.

We also compared the effects of nitrate leaching reduction among the three patterns for a reduction of 50 kg ha $^{-1}$ in N input at five application rates: 300, 250, 200, 150, and 100 kg ha $^{-1}$. In all 50 kg ha $^{-1}$ reduction scenarios, the N leaching reductions of the IPCC and 19% model estimates were 15 and 9.5 kg NO $_3$ -N ha $^{-1}$, respectively. When the N rate was decreased from 300 to 250 kg ha $^{-1}$, the reduction in ΔEF to 9.75 kg NO $_3$ -N ha $^{-1}$ was 35% lower than that of the IPCC model estimate, but 3% higher than that of the Lin et al. (2001) 19% EF model. When the N rate was decreased from 200 to 150 kg ha $^{-1}$, the N leaching reductions for the ΔEF pattern were 55% and 29% less than the IPCC model and the 19% EF estimates, respectively. In comparison with other linear models, our ΔEF model predicted only a relatively small reduction in nitrate leaching when N input reductions were reduced. For example, when N input was reduced from 100 to 50 kg ha $^{-1}$, our ΔEF model calculated a leaching reduction of only 3.69 kg NO $_3$ -N ha $^{-1}$, which was smaller than the reductions calculated by the IPCC and 19% models.

The N input levels for maize and wheat cropping systems in major countries were calculated from the N fertilizer inputs provided by the IFA and arable land area information in the FAO database (Table S1). All countries and crops were divided into four N input categories: $0 < N \leq 50$, $50 < N \leq 100$, $100 < N \leq 150$, and $N > 150$, containing data for 10, 16, 9, and 11 country-crop combinations, respectively. Differences in the mean N application rates and nitrate leaching per hectare (kg NO $_3$ -N ha $^{-1}$) evaluated by the three different leaching patterns are listed in Tables 2 and S2. Due to the low levels of N input in most crop-country combinations ($0 < N \leq 50$ and $50 < N \leq 100$; $n = 10$ and $n = 16$, respectively), the ΔEF pattern yielded a smaller N leaching value than the other two patterns (Fig. 3B; Table 2). Comparing the mean worldwide N leaching values calculated using the three patterns, the mean ΔEF result was 8.04 kg NO $_3$ -N ha $^{-1}$, which is 61% less than the estimate of the Lin et al. (2001) model (20.74 kg NO $_3$ -N ha $^{-1}$), and 75% less than the IPCC model (32.74 kg NO $_3$ -N ha $^{-1}$) (Table 2).

4. Discussion

Across nations, the constant EF estimation did not explain why N fertilizer-induced nitrate leaching varied with N application rate. This failure to provide an explanation was especially marked under conditions in which N leaching increased dramatically when the availability of soil N exceeded crop requirements (Lord and Mitchell, 1998; Perego et al., 2012; Delin and Stenberg, 2014). Our global meta-analysis was performed to address the difficulties encountered by linear

Table 2

Estimates of nitrate leaching produced by different EF models for wheat and maize crop-lands in major countries. N fertilizer input and arable land area data for maize and wheat were obtained from the International Fertilizer Industry Association (IFA) and the Food and Agriculture Organization (FAO), respectively. Countries with no available data were excluded from the calculations (see Table S1). EF, soil nitrate leaching emission factor.

N rate (kg N ha $^{-1}$)	Country-crops	Mean (kg N ha $^{-1}$)	N leaching per hectare (kg NO $_3$ -N ha $^{-1}$)		
			IPCC	19% Model	ΔEF
$0 < N \leq 50$	10	33.33	10.00	6.33	1.29
$50 < N \leq 100$	16	78.56	23.57	14.93	4.17
$100 < N \leq 150$	9	131.12	39.33	24.91	8.99
$N > 150$	11	204.53	61.36	38.86	19.00
Total	46	109.13	32.74	20.74	8.04

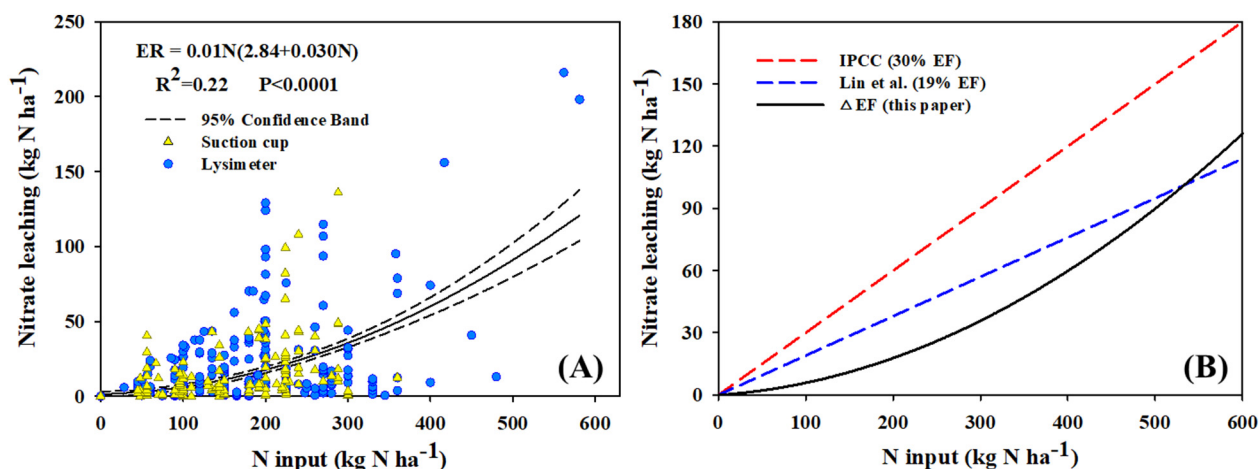


Fig. 3. (A) Quadratic response (with 95% confidence intervals) of nitrate leaching to N application rate on arable uplands. Triangles and circles indicate data obtained by the suction cup and lysimeter methodologies, respectively. (B) Comparison of nitrate leaching estimates provided by the International Panel on Climate Change (IPCC; 30% EF), Lin et al. (2001; 19% EF), and the Δ EF calculated in the current meta-analysis ($0.01 N [2.84 + 0.030 N]$). EF, soil nitrate leaching emission factor; ER, the amount of nitrate leaching; N, nitrogen application rate.

modeling of the nonlinear response of soil nitrate leaching to increasing N fertilization. We found a positive mean Δ EF for all site-year combinations and most crop categories, N fertilizer types, and other environmental or management factors (Table 1). The main results of this study indicate that fertilizer-induced EFs of nitrate leaching are not constant but increase with increasing N application rates; thus, nitrate leaching in response to changes in N inputs exceeds the commonly accepted linear relationship. Our results are consistent with previous findings for nitrous oxide and ammonia emissions related to multiple N inputs (Shcherbak et al., 2014; Hoben et al., 2011; Jiang et al., 2017; Chen et al., 2014).

Among the main crop types, rice had the lowest Δ EF value, perhaps because N leaching in the rice season was much lower than leaching in upland croplands. An experiment conducted in China showed that the N leaching EF from a flooded rice field was only 2.0–2.7% (Zhu and Chen, 2002). As N application levels increase, the EF maintains low-level results at smaller Δ EF values. Less NO_3^- leaching occurs during the rice growing season than in upland crops due to the lower nitrification capacity of the soil; ammonia volatilization then becomes the major N loss pathway (Zhao et al., 2009). Conversely, vegetable crop production had a higher Δ EF value than other crop categories, contributing to low-efficiency irrigation, frequent cultivation, and especially excessive N input that surpassed crop requirements, thereby making vegetable production vulnerable to nitrate leaching (Jiang et al., 2012; Zhang et al., 2017; Wang et al., 2018).

Ammonium nitrate (AN) had a significantly higher Δ EF for N leaching than any other fertilizer type, which is consistent with the result of Shcherbak et al. (2014). Fan and Li (2009) conducted a field experiment to examine the relationship between fertilizer type and nitrate leaching. Similar to our results, they found that AN was a very soluble fertilizer and that N leaching reached high levels within a short period of time Fan and Li (2009). In our study, the nitrate leaching Δ EF for CRF was low, but similar to that of other fertilizer types, and the CRF Δ EF was still positive (Table 1). Previous work also suggested that CRF increased the nitrate concentration in soil relative to unfertilized soils, thus increasing the risk of N leaching; nevertheless, CRF may improve N use efficiency (Zhang et al., 2018; Ke et al., 2018).

We also showed that the effects of SOC and TN on Δ EF were highest when SOC and TN values were high. The high soil organic matter associated with high TN may increase the N soil pool, and accelerate mineralization and nitrate leaching (Di and Cameron, 2002; Gai et al., 2019). Our pH results, which showed that acidic soil conditions yielded a larger Δ EF and thus a greater amount of nitrate leaching, were inconsistent with previous reports indicating that partial alkaline conditions are beneficial to nitrification, thus increasing the nitrate concentration in the

soil solution (Boer and Kowalchuk, 2001; Wang et al., 2019). This discrepancy may be explained by specific experimental conditions (such as fertilizer application or soil texture) leading to a higher EF; this possibility remains to be verified.

Higher Δ EF values occur when there is more precipitation; rainfall is a determining factor in nitrate leaching (Liang et al., 2011). When water supply is insufficient, N accumulates in soil, mainly in the root layer. Once the water supply exceeds the soil field capacity, nitrate washes out from the rhizosphere and leaches into the groundwater. Δ EF values were higher when the fertilizer was applied once, but with multiple fertilization applications, fertilizer-induced nitrate leaching was relatively high. This result can be explained by conditions during the early growth stages when the soil nitrate concentration is high but the crop N demand is too low for significant uptake; the redundant nitrate capacity increases the risk of leaching (Xia et al., 2017; Lu et al., 2019).

Our Δ EF model did not reproduce the results of the IPCC 30% EF and 19% EF (Lin et al., 2001) models across a fertilizer application range of 0–500 kg ha⁻¹ N. Our nitrate leaching estimates indicate that the IPCC fertilizer-induced 30% EF and 19% EF models may produce overestimates for upland crops (Fig. 3B). Another recent global meta-analysis based on 205 observations demonstrated that, on average, 22% and 15% of applied fertilizer was lost by nitrate leaching during maize and wheat growth, respectively (Zhou and Butterbach-Bahl, 2014). The high N leaching estimates in previous studies might be explained in part by the fact that many site-year studies did not include a control and did not subtract the nitrate leaching in the control from leaching in the N fertilization treatments. Many recent top-down evaluations have shown great variability in N leaching due to diverse management and environmental conditions (Quemada et al., 2013; Cui et al., 2014; Eagle et al., 2017). For example, an analysis of wheat and maize farming found that high-yield lands had extremely high N leaching EF values of 21% and 25%, respectively. However, highly efficient farms had average values of only 7% and 13% EF values for wheat and maize, respectively (Cui et al., 2014); these lower values are close to our estimates. Long-term experiments may detect the average effect of N leaching induced by fertilizer application. Goulding et al. (2000) provided a strong case for the long-term experiment conducted at Rothamsted, UK: averaging over 8 years, 9% of applied N was leached in the Broadbalk Experiment (Goulding et al., 2000). This result was consistent with our Δ EF estimate; we obtained a 9% EF value at an N application rate of 200 kg ha⁻¹.

Other recent reports have shown that the response curve of nitrate leaching with increasing N inputs rises rapidly when the N input exceeds the optimum, indicating that a nonlinear solution will provide a more robust estimator of accurate nitrate leaching inventories than a constant linear analysis. Delin and Stenberg (2014) described the

response using the exponential function $y = y_0 + 1.4 \exp(0.027x)$ ($R^2 = 0.84$; $P = 0.001$), and demonstrated that fertilization significantly affected nitrate leaching at high N levels in Sweden. Moreover, Zhou and Butterbach-Bahl (2014) simulated the N leaching response curve of wheat using the nonlinear regression expression $y = 16.86 \exp(0.003x)$ ($R^2 = 0.17$; $P < 0.001$). The exponential response often overestimated nitrate leaching at low N application rates (in comparison with our ΔEF model), but the results were more consistent between models when the N input exceeded 300 kg N ha^{-1} . Zhou and Butterbach-Bahl's (2014) response curve estimated nitrogen leaching value of $22.42 \text{ kg NO}_3\text{-N ha}^{-1}$ (24% of applied N) at a N application rate of 95 kg N ha^{-1} (the global average N rate for wheat; Table S1), whereas our ΔEF model estimate was only $5.43 \text{ kg NO}_3\text{-N ha}^{-1}$.

Worldwide agricultural N balances differ substantially with agricultural and social development, from inadequate inputs in parts of many developing countries to excessive and environmentally damaging surpluses in many developed and rapidly growing countries (Vitousek et al., 2009). These N imbalances highlight the global implication of a nonlinear EF response for global nitrate leaching inventories. High nitrate leaching induced by N fertilizer application resulted in N hotspots with high reactive N losses (Gu et al., 2012; Mason et al., 2017). This occurs in regions where agricultural intensification and fertilizer application rates are high, including China and India, and active N losses to the environment are consequently high. Non-hotspot regions with low N inputs include those in sub-Saharan Africa (e.g., Malawi; Zhang et al., 2015), where agricultural intensification and social development indices are relatively low, and little chemical fertilizer is devoted to agricultural production (Zhang et al., 2015; Vitousek et al., 2009).

When different countries regulate their N inputs, the risk of nitrate leaching and environmental cost will change in a non-linear manner. Our ΔEF model is expected to increase the accuracy of scenario analyses and assessments of the effects of changing fertilizer inputs on nitrate leaching across countries with different N inputs. Numerous field tests have demonstrated that an appropriate reduction in N input will not significantly affect crop yields in countries with high N input. For example, Ying et al. (2017) confirmed that when the N rate is reduced from 279 to 175 kg N ha^{-1} , wheat yield remains stable, resulting in significantly increased ecological and social benefits on the North China Plain. High N-input countries have made an effort to control fertilizer consumption, resulting in policies such as the 'Zero Increase Action Plan' promoted by the Ministry of Agriculture of China to control increases in fertilizer use by 2020 (Liu et al., 2015). The reduction in cumulative nitrate leaching through optimizing N management in hotspot regions will be underestimated by the constant EF model due to nonlinear responses. In contrast, in developing regions such as sub-Saharan Africa, where N inputs are low, the increase in cumulative nitrate leaching and environmental costs will be overestimated by a constant EF (relative to our ΔEF model). These projections are important because increasing N application rates will improve crop production, and social and agricultural development in poorly developed regions of the planet.

5. Conclusion

We conducted a global meta-analysis of 324 site-year combinations to quantify the nonlinear response of nitrate leaching induced by N fertilizer application. We found that ΔEF was positive and significantly different from zero, indicating that EF increases monotonically, but not at a constant rate, in response to N fertilizer application. Thus, variation in nitrate leaching exceeds the commonly accepted linear relationship. Furthermore, a comparison of ΔEF model results with results of two other previously reported constant EF values provided insight into a more accurate method for estimating N leaching and potential environmental costs induced by N fertilizer application. Future research should include expanded controlled field experiments testing multiple

independent variables and the development of more accurate models for application under diverse cropland conditions.

Author contributions

Z.L.C. designed the study; Y.C.W., H.Y., Y.L.Y. and H.F.Z. performed the research; Y.C.W. and H.Y. analyzed the data; Y.C.W. wrote the paper. All authors critically reviewed the manuscript.

Conflicts of interest statement

The authors declare no competing financial interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.12.029>.

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